

SPECIFICATION AMENDMENTS

Please replace paragraph [0018] in the specification on page 6 of the application with the following paragraph:

[0018] European Patent Publication No. EP 1,118,370 A1 describes a PSA process with ten or more adsorbent beds that utilizes four equalization steps that can be used for hydrogen purification. The cycles described are effective for achieving high hydrogen recovery at three atmospheres and 74% hydrogen in the PSA feed gas. Cycles with large numbers of equalization steps are more efficient at higher pressure, but at lower pressures they add more complexity to the PSA process without providing a significant performance benefit.

Please replace paragraph [0019] in the specification on page 6 of the application with the following paragraph:

[0019] International Patent Publication No. WO 02/04096 A1 discloses a process for purifying a hydrogen stream containing carbon monoxide and nitrogen using a Ca exchange X-zeolite at pressures between 20 and 50 bara. U.S. Patent No. 6,302,943 discloses a PSA process for purifying hydrogen from a gas stream containing carbon monoxide and/or nitrogen using a PSA system operating between 100 and 1,000 psig. U.S. Patent No. 6,027,549 discloses [[s]] a PSA process using an activated carbon with a density between 35 and 38 lb/ft³ for removing carbon dioxide from hydrogen in a PSA process operating above 350 psig. U.S. Patent No. 5,912,422 discloses the use of a lithium-exchange faujasite in a PSA process to remove carbon monoxide from a feed stream containing hydrogen at pressures between 5 and 70 atmospheres. In this process, a first adsorbent is used for removal of carbon dioxide and C₁ – C₈ hydrocarbons.

Please replace paragraph [0021] in the specification on page 7 of the application with the following paragraph:

[0021] U.S. Patent No. 5,753,010 describes a PSA process where a portion of a depressurization and purge of fluent effluent gases from a PSA system is repressurized and then recycled back to the PSA system in order to increase product recovery. This process, which runs at a pressure of at about 150 psig, requires significant energy for compression, not only for the feed gases to reformer, but also for repressurizing the depressurization and purge of fluent effluent gases from the PSA system.

Please replace paragraph **[0022]** in the specification on page 7 of the application with the following paragraph:

[0022] Some gas purifying processes combine membrane and PSA processes in a hydrogen purification system. U.S. Patent No. 4,863,492 describes a process where the reformate stream is first fed to a membrane separator to generate a hydrogen rich permeate. The permeate is then fed to a PSA unit to further purify the hydrogen. The feed gas applied to the combined membrane/PSA hydrogen purification system is at least 200 psig. U.S. Patent No. 4,398,926 discloses a similar process, but where a portion of the membrane retinate retentate is co-fed to the PSA system along with a permeate to improve hydrogen recovery. This process is designed for feed pressures above 600 psig.

Please replace paragraph **[0027]** in the specification on page 8 of the application with the following paragraph:

[0027] Figure 2 is an illustration of a PSA cycle for the 9-bed PSA system shown in figure Figure 1, according to an embodiment of the present invention;

Please replace paragraph **[0033]** in the specification on page 9 of the application with the following paragraph:

[0033] Figure 8 is a stationary port plate used in combination with the rotary feed valve shown in figure Figure 7;

Please replace paragraph **[0035]** in the specification on page 9 of the application with the following paragraph:

[0035] Figure 10 is a plan view of a stationary port plate used in combination with the rotary product valve shown in figure Figure 9.

Please replace paragraph **[0037]** in the specification on page 9 of the application with the following paragraph:

[0037] Figure 1 is a plan view of a PSA system 10 for purifying a feed gas, such as a reformate gas, into a product gas, such as a purified hydrogen gas. The system 10 includes nine columns, beds or vessels 12 each having a feed end 14 that receives the feed gas and a product end 16 that emits the product gas. The vessels 12 include an adsorbent or mixture of adsorbents for adsorbing carbon monoxide, carbon dioxide, nitrogen, water, methane, etc. At least one adsorbent in the vessels 12 is either zeolite 5A or zeolite LiX. A combination or a mixture of zeolite 5A and zeolite LiX can also be used. Additionally, a second adsorbent, such as activated carbon, activated alumina, zeolite 13X or zeolite 4A can be used at the feed end 14 of the vessels 12 for adsorbing some of the water and carbon dioxide in the feed gas. The adsorbents in the vessels 12 will also remove other impurities that may be in the feed gas, such as heavier hydrocarbons like ethane, propane, butane, ethylene, propylene, hydrogen sulfide and/or NH₃.

Please replace paragraph **[0045]** in the specification on page 12 of the application with the following paragraph:

[0045] Table I below shows a PSA cycle, according to the invention, for the system 10 that depicts the sequencing of the valves 28. The number on the top column is a particular cycle period, and the identification of the valve 28 below the number shows that that valve 28 is open. Figure 2 is an illustration of the PSA cycle depicted in Table I. Eighteen cycle periods are shown in figure Figure 2 and Table I because each PSA cycle for a particular vessel 12 requires twice as many cycle periods as there are number of the vessels 12.

Please replace paragraph **[0046]** in the specification on page 12 of the application with the following paragraph:

[0046] The stages of the PSA cycle of Table I and figure Figure 2 can be described as follows. As can be seen from figure Figure 2, the cycle steps described below are performed in the following sequence. During a production stage of a particular vessel 12, the feed gas is delivered from the feed manifold 20 to the feed end 14 of the vessel 12 through the corresponding feed valve VF, and the product gas is drawn from the product end 16 of the vessel 12 into the product manifold 22 through the corresponding product valve VP. The production stage lasts six cycle periods for each vessel 12.

Please replace paragraph **[0047]** in the specification on pages 12 and 13 of the application with the following paragraph:

[0047] During a first equalization down (E1 down) stage of a particular vessel 12, the product end 16 of the particular vessel 12 is connected to the product end 16 of an adjacent vessel 12 that is at a lower equalization pressure through the corresponding equalization valve VQ. The E1 down stage lasts for one cycle period. For the discussion below, the particular vessel 12 can be bed 1, the adjacent vessel 12 can be bed 2 and the other adjacent vessel 12 can be bed 9 with reference to figure Figure 2. However, it will be understood by those skilled in the art, that all of the vessels 12 in the system 10 go through the same cycle. The adjacent vessel 12 completed a second equalization up (E2 up) stage at the previous cycle time period, described below. The adjacent vessel 12 undergoes a first equalization up (E1 up) stage,

described below, while the first vessel 12 is in the E1 down stage. The pressure in the vessel 12 is lowered from the production pressure to the higher equalization pressure during the E1 down stage.

Please replace paragraph [0056] in the specification on page 15 of the application with the following paragraph:

[0056] A PSA cycle for the system 10 can be employed that has three or four equalization stages. The PSA cycle for a 9-bed PSA system with three equalization stages is illustrated in figure Figure 3. In order to accommodate for the extra equalization stage, the duration of the purge stage is reduced by two cycle periods from the PSA cycle shown in figure Figure 2. This is because the added E3 down stage and the E3 up stage each require a single cycle period. The advantage of a PSA cycle with more equalization stages would be a higher hydrogen product recovery. However, more valves 28 would be required as more equalization steps are added. For example, a 9-bed PSA system operating with a three equalization stage PSA cycle would have fifty-four valves 28, and would also need an additional manifold for equalization gas flow.

Please replace paragraph [0057] in the specification on pages 15 and 16 of the application with the following paragraph:

[0057] Similarly, a PSA cycle for a 9-bed PSA system with four equalization stages would have an even shorter purge stage, as illustrated in figure Figure 4. Effective purging of the adsorbent could still be accomplished, even in a shorter time, by increasing the amount of purge gas allowed through the reduction valve 30. A 9-bed PSA system operating with a four-equalization stage PSA cycle would have sixty-three valves 28, and two additional manifolds for equalization gas flow.

Please replace paragraph [0058] in the specification on page 16 of the application with the following paragraph:

[0058] The PSA system 10 of the invention is not specifically limited to 9-beds. It is recommended that the system 10 have at least five adsorbent vessels 12 to enable at least two equalization stages. A PSA cycle for a 5-bed PSA system with two equalization stages is illustrated in figure Figure 5. A fixed-bed PSA system operating with this PSA cycle would need twenty-five valves 28, and thus, would be a simpler system than the 9-bed PSA system 10 described above. However, the productivity of the 5-bed PSA cycle is lower than that of the 9-bed PSA cycle as the percentage of the cycle that each bed is in the production stage is lower in the 5-bed system (2 of 10 time steps or 20%) than in the 9-bed system (6 of 18 time steps or 33.3%). Thus, the choice of the optimal number of beds used in the PSA cycle is a trade off between performance, i.e. recovery and productivity, versus cost and complexity.

Please replace paragraph **[0059]** in the specification on page 16 of the application with the following paragraph:

[0059] The cycle for a PSA system with more than nine vessels 12 can also be used. For example, a 12-bed PSA system with three-equalization stages, 33.3% of the cycle as production, and equal production and purge stage duration is illustrated in figure Figure 6.

Please replace paragraph **[0066]** in the specification on page 18 of the application with the following paragraph:

[0066] When an aperture 50 is fully covered by the face 56 of the feed valve 40, no gas flows into or out of the feed end 14 of the corresponding vessel 12. This situation exists during the equalization and product pressurization stages of the PSA cycle. At any given time, either two or three of the apertures 50 are fully covered by the face 56 of the feed valve 40. The feed valve 40 rotates counter clockwise along the axis 52 relative to the feed port plate 48 to generate the PSA cycle described above and shown in figure Figure 2. Each rotation of the feed valve 40 is equivalent to two of the PSA cycles shown in figure Figure 2.

Please replace paragraph [0067] in the specification on pages 18 and 19 of the application with the following paragraph:

[0067] Figure 9 is a plan view of a rotary product valve 68 that can replace all of the product valves VP1 – VP9, the purge valves VR1 – VR9 and the equalization valves VQ1 – VQ9 in the system 10. Figure 10 is a plan view of a stationary product port plate 70 used in combination with the product valve 68. The port plate 70 includes nine product apertures 72, one for each vessel 12, that are evenly spaced (40° apart) at the same radial distance from a center axis 78. The apertures 72 are connected to the product end 16 of each of the nine vessels 12 by a suitable conduit (not shown). A face 76 of the product port plate 70 is lapped flat to within 20 millionths of an inch of a face 80 of the valve 68. The product valve 68 is aligned with the axis 78 so that its face 80 is in direct contact and completely flush with the face 76 of the feed product port plate port 70.

Please replace paragraph [0069] in the specification on page 19 of the application with the following paragraph:

[0069] The valve 68 includes eight equalization ports 88-102 88, 90, 92, 94, 96, 98, 100 and 102. The equalization ports 88 and 102, the equalization ports 90 and 92, the equalization ports 94 and 96 and the equalization ports 98 and 100 are spaced 10° apart. The equalization ports 88 and 90, the equalization ports 92 and 94, the equalization ports 96 and 98 and the equalization ports 100 and 102 are spaced 80° apart. The equalization ports 88 and 90, the equalization ports 92 and 94, the equalization ports 96 and 98, and the equalization ports 100 and 102 are connected to each other through discrete conduits within the valve 68. The equalization ports 88-102 88, 90, 92, 94, 96, 98, 100 and 102 are wide enough to just cover one product aperture 72. At any given time, two equalization ports 88-102 88, 90, 92, 94, 96, 98, 100 and 102, which are connected to each other, i.e., ports 88 and 90, are exchanging gas with two product apertures 72. The other six equalization ports 88-102 88, 90, 92, 94, 96, 98, 100 and 102 are fully covered by the face 76 of the product port plate 70.

Please replace paragraph [0073] in the specification on page 20 of the application with the following paragraph:

[0073] The product valve 68 rotates clockwise on the axis 78 relative to the product port plate 70 to generate the PSA cycle described above and shown in figure Figure 2. Each rotation of the product valve 68 is equivalent to two PSA cycles as described in figure Figure 2. The feed valve 40 and the product valve 68 rotate about the same center axis 78 at the same speed and, therefore, are driven by a single common motor.

Please replace paragraph [0074] in the specification on page 21 of the application with the following paragraph:

[0074] Rotary valves and stationary port plates could readily be designed for any PSA cycle, including those shown in figures Figures 3-6. For the 9-bed cycles illustrated in figures Figures 3 and 4, different feed and product valves only need to be designed, as opposed to the addition of one on/off valve per bed for each additional equalization stage in the PSA system 10 illustrated in figure Figure 1. Different feed and product port plates would be necessary depending on the number of the vessels in the PSA system 10, with the port plates having one aperture for each vessel 12.

Please replace paragraph [0078] in the specification on page 22 of the application with the following paragraph:

[0078] In one example, a 9-bed PSA system was equipped with the rotary valves and port plates shown in figures Figures 7-10 and used the PSA cycle illustrated in figure Figure 2. The adjustable screws 108 in the product valve 68 were set so that 7 SLPM of nitrogen would flow through each purge gas port 110 when 30 psig of nitrogen was applied to the product manifold 22 and there was no back pressure down stream of the purge gas ports 110. Each of the vessels 12 was filled with 378g of UOP 5A-MG

adsorbent. 280 SLPM of reformate (47.4% hydrogen, 1.0% carbon monoxide, 15.2% carbon dioxide, 5.9% water, and 30.5% nitrogen), at 35.7 psig and 70° C was fed into the feed manifold 20 of the PSA system 10. The force at which the valves 40 and 68 and the port plates 48 and 70 are compressed together was optimized for these operating conditions. The valves 40 and 68 were rotated at 6.1 rpm, generating a PSA cycle time of 4.9s. The product gas collected from the PSA system 10 at steady state is summarized in table II below. 70% of the hydrogen in the feed gas is recovered, which is unexpectedly high for a PSA system operating at 35 psig. Also, the product carbon monoxide is within acceptable levels for PEM fuel cell operation.

Please replace paragraph [0079] in the specification on page 23 of the application with the following paragraph:

[0079] In another example, a 9-bed PSA system equipped with the rotary valves 40 and 68 and the port plates 48 and 70, shown in figures Figures 7-10, and used the PSA cycle depicted in figure Figure 2. The adjustable screws 108 in the product valve 68 were set in the same position as in the example discussed above. The force at which the valves 40 and 68 and the port plates 48 and 70 are compressed together was also the same as in that example. Each of the vessels 12 was filled with 378g of UOP 5A-mg adsorbent. 280 SLPM of reformate gas, 47.4% hydrogen, 1.0% carbon monoxide, 15.2% carbon dioxide, 5.9% water and 30.5% nitrogen, at 45.2 psig and 70° C was fed into the feed manifold 20 of the PSA system 10. The valves 40 and 68 were rotated at 3.8 rpm, generating a PSA cycle time of 7.9 seconds. The product gas collected from the PSA system 10 at steady state is summarized in table III below. Over 73% of the hydrogen in the feed gas is recovered. There is also a four fold reduction in the product carbon monoxide, resulting from the relatively small increase in pressure from the example discussed above. It is reasonable to expect that the recovery could be improved further by optimizing the purge gas flow rate by adjusting the adjustable screws 108 and by optimizing the force at which the valves 40 and 68 and the port plates 48 and 70 are compressed together.